

IN THE CLAIMS

Claims 1-2. (Cancelled)

Claim ²~~3~~. (Allowed) The process according to claim 22, wherein said non-hydrolyzable substituent is H, an alkyl, aryl or fluoroalkyl group or an aminoalkyl group.

Claim ³~~4~~. (Allowed) The process according to claim ¹~~22~~, wherein said step c) of drying the gel is a liophilisation carried out at a pressure lower than 70 mm Hg, to obtain a mesoporous aerogel powder.

Claim ⁴~~5~~. (Allowed) The process according to claim ¹~~22~~, wherein said step c) of drying the gel is a mild heat treatment carried out at an atmospheric pressure and a temperature no greater than 100°C.

Claim ⁵~~6~~. (Allowed) The process according to claim ¹~~22~~, wherein said nitroxyl radical is added to said solution along with said monomer precursor in a one-step procedure.

Claim ⁶~~7~~. (Allowed) The process according to claim ¹~~22~~, wherein in a two-step procedure, first said monomer precursor is hydrolyzed in part with water in the

presence of an acid and then said nitroxyl radical is added to this solution, to obtain a porous sol-gel polymeric oxide with a fractal macromolecular structure.

Claim 8. (Cancelled)

⁷
1 Claim ~~9~~ (Allowed) The process according to claim ~~22~~, wherein said reductive amination is carried out by stirring a solution of said 4-oxo-TEMPO in methanol with said 3-aminopropyl-trimethoxysilane, and reducing the thus formed imine with NaBH_3CN .

⁸
5 Claim ~~10~~ (Allowed) The process according to claim ~~8~~, wherein said nitroxyl radical is TEMPO or a precursor thereof and said radical is physically entrapped within a sol-gel matrix adding a solution thereof in methanol to said precursor following said one-step procedure.

⁹
1 Claim ~~11~~ (Allowed) The process according to claim ~~22~~, wherein said catalytic porous materials are in the shape of powders, films, monoliths, or fibers.

Claims 12-13. (Cancelled)

¹¹
Claim ~~14~~. (Allowed) A process according to
¹⁰
claim ~~23~~, wherein said liquid phase is an organic
solvent, a biphasic organic solvent-water system, or
water and said primary oxidant is NaOCl, NaOBr, HNO₃,
CuCl/O₂, K₃Fe(CN)₆, or NO₂.

¹²
Claim ~~15~~. (Allowed) A process according to
¹⁰
claim ~~23~~, wherein said alcohol substrate is an alkyl
alcohol, an aryl alcohol, a steroid alcohol, an allylic
alcohol, a terpenoid alcohol or retinol and it is
oxidized in a bi-phasic reaction system CH₂Cl₂-H₂O, said
primary oxidant is aqueous alkaline NaOCl and wherein
said nitroxyl radical is 4-oxy-TEMPO and said monomer
precursor is 3-aminopropyl-trimethoxysilane to obtain a
catalytic material containing chemically linked radicals.

¹³
Claim ~~16~~. (Allowed) A process according to
¹⁰
claim ~~23~~, wherein said alcohol substrate is a monomer or
an oligomeric carbohydrate protected at the anomeric
center, said solvent is water, said oxidant is alkaline
NaOCl or NaOCl in the presence of a catalytic amount of
NaBr, and wherein said nitroxyl radical is 4-oxy-TEMPO
and said monomer precursor is 3-amino-propyl-
trimethoxysilane to obtain a catalytic material
containing chemically linked radicals.

¹⁴
Claim ~~17~~. (Allowed) A process according to
claim ¹³~~16~~, wherein said catalytic material is in the form
of pumice stones coated with said sol-gel film doped with
said nitroxyl radicals, and said carbohydrate is a water
soluble polymer.

¹⁵
Claim ~~18~~. (Allowed) The catalytic material
doped with a chemically linked nitroxyl radical obtained
with a process as claimed in claim ~~22~~.

¹⁶
Claim ~~19~~. (Currently amended) The process
according to claim ~~1~~ ¹~~22~~, wherein P is a non-hydrolyzable
substituent.

Claim 20. (Cancelled)

¹⁷
Claim ~~21~~. (Allowed) A process according to
claim ¹⁰~~28~~, wherein said alcohol substrate is an alkyl
alcohol, an aryl alcohol, a steroid alcohol, an allylic
alcohol, a terpenoid alcohol or retinol and it is
oxidated in a bi-phasic reaction system $\text{CH}_2\text{Cl}_2\text{-H}_2\text{O}$,
wherein said primary oxidant is aqueous alkaline NaOCl ,
and wherein said nitroxyl radical is 4-oxy-TEMPO and said
monomer precursor is 3-aminopropyl-trimethoxysilane to

obtain a catalytic material containing chemically linked radicals, wherein said radical is tethered to said monomer precursor through reductive amination by stirring for three hours a solution of 4-oxo-TEMPO in methanol with a slight excess of 3-aminopropyl-trimethoxysilane, and reducing the thus formed imine with NaBH_3CN .

Claim ~~22~~. (Allowed) A process for the preparation of a reactive sol-gel catalytic porous material comprising chemically doping said material with stable organic nitroxyl radicals, by carrying out the steps of:

copolymerizing a solution including:

a) 3-amino-propyl-trimethoxysilane as a monomer precursor;

b) a dopant consisting of 4-oxy-TEMPO as a stable nitroxyl radical or a precursor thereof;

c) a solvent including H_2O and a co-solvent selected from the aliphatic alcohols; an acid or base to catalyze the processes of sol-gel hydrolysis and copolymerization; and one or more additives selected from those known to be useful in the preparation of porous materials to form a gel containing said dopant trapped therein;

d) tethering said radical to said monomer precursor through reductive amination, said solution including H₂O as a solvent and a co-solvent from the aliphatic alcohols; an acid or base to catalyze the processes of sol-gel hydrolysis and copolymerization; and one or more additives selected from those known to be useful in the preparation of porous materials to form a gel containing said dopant chemically trapped therein;

e) evaporating said solvent;

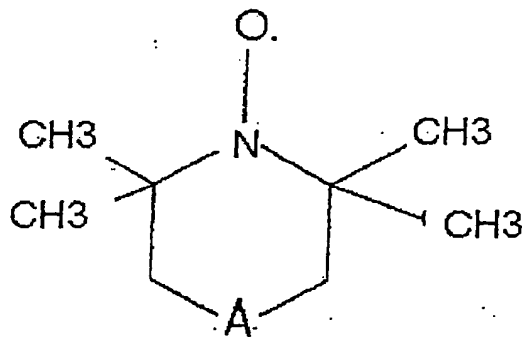
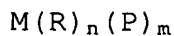
f) drying said gel; and

g) coating said gel on a mesoporous inorganic support.

¹⁰
Claim 23. (Allowed) A process for a liquid-phase oxidative conversion of a substrate of a primary or secondary alcohol into a carbonyl or carboxyl derivative thereof, comprising said conversion being carried out with said substrate in catalytic presence of a reactive sol-gel catalytic porous material either chemically or physically doped with stable organic nitroxyl radicals, said material being coated on a mesoporous inorganic support and containing a copolymer of

a) at least one monomer precursor selected from the group consisting of metal and semi-metal alkoxides,

metal esters and semi-metal esters, of the general formula



wherein M is a metal or a semimetal, R is an hydrolyzable substituent, P is a non-hydrolyzable group, n is an integer of 1 to 6, and m is an integer of 0 to 6, and

b) a dopant consisting of a stable di-tertiary-alkyl nitroxyl radical or a precursor thereof of formula, wherein A represents a chain of two or three carbon atoms, one or two of said carbon atoms being eventually substituted by one oxygen or nitrogen atom, and one or more additives selected from those known to be useful in the preparation of porous materials to form a gel containing said dopant trapped therein.

Claim ¹⁶~~24~~. (New) A process for liquid-phase oxidative conversion of a substrate of a primary or secondary alcohol into a carbonyl or carboxyl derivative thereof, comprising conducting said oxidative conversion

in the presence of a doped catalytic material according
to claim ~~22~~¹, and in the presence of a primary oxidant
effected in selective alcohol oxidations mediated by
nitroxyl radicals.